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Induced Magnetic Moment on Iridium in Ir-Doped Chromium Dioxide Probed by Circular Magnetic Dichroism

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Abstract. We report X-ray Magnetic Circular Dichroism (XMCD) measurements performed at the $L_{II,III}$ edges of Ir in $Cr_{0.975}Ir_{0.025}O_2$. The data were recorded in transmission mode at beamline 6 of the European Synchrotron Radiation Facility. They allow us to determine the z projection of the orbital angular momentum of the $5d$ electrons of Ir. The results obtained by the XMCD technique at the Ir edges and ^{193}Ir Mössbauer spectroscopy are compared.

CrO_2, a half - metallic ferromagnet (T_C = 398 K), is well known for its use in magnetic recording applications, owing to its suitable remanent magnetization and coercivity which are extrinsic properties of this material. Substitutions of Cr by Ir, even with small amounts (a few percent), increase notably the coercitive field (150 - 200 mT) compared with that of the pure material (5 - 50 mT) [1, 2]. The role of the spin orbit coupling of Ir ($\xi \simeq 3000$ cm^{-1}) has been underlined [1, 2] and recent band structure calculations of the $Cr_{1-x}Ir_xO_2$ solid solutions have shown that the orbital to spin magnetic moment ratio (μ_L/μ_S) is 8 times larger for Ir than for Cr [3]. Another prediction from these calculations was that the coupling between the Cr and Ir spin moments (μ_S^{Cr} and μ_S^{Ir}) is antiparallel and the Ir orbital magnetic moment (μ_L^{Ir}) is parallel to μ_S^{Ir} (see Fig. 1). In order to shed more light on the effects of Ir substitutions within the lattice of CrO_2, we investigated the microscopic magnetic behaviour of Ir by using ^{193}Ir Mössbauer effect measurements and X-ray Magnetic Circular Dichroism (XMCD) experiments at the $L_{II,III}$ edges of Ir in $Cr_{0.975}Ir_{0.025}O_2$. The particles with this composition were prepared by submitting the precursors of the solid solution to very high oxygen pressure treatments using $KClO_3$ as oxygen source in a belt type apparatus [1].

The ^{193}Ir Mössbauer spectrum of $Cr_{0.975}Ir_{0.025}O_2$ recorded at 4.2 K using conventional methods [4] was accounted for assuming a combined magnetic and quadrupolar interactions with a hyperfine field $H_{hf}^{1/2} = (-)$ 67.1 (1.0) T and a quadrupolar coupling constant $e^2qQ = -2.86$ (2) mm/s. The isomer shift of -0.13 (3) mm/s vs Ir metal characterizes the stabilization of Ir^{4+} ions (d^5) within the CrO_2 lattice. Of special interest is the value of the hyperfine anomaly $^{3/2}\Delta^{1/2} = 0.092$ (2) because it allows to separate out the different contributions to the hyperfine field [4]. $H_{hf} = H_C + H_L + H_{SD}$ where H_C, H_L and H_{SD} are respectively the core polarization, orbital and spin dipolar fields. Neglecting the last contribution, the analysis of H_{hf} leads to $H_L = -2\mu_B\langle r^{-3}\rangle_{5d}\langle L_z\rangle = 31$ (13) T and $H_C = -98$ (12) T. Taking $\langle r^{-3}\rangle_{5d}$ 12.455 a.u. and $H_C/2\langle S_z\rangle = 60$ (3) T [5] we obtain $\langle L_z\rangle = -0.20$ (8) and $\langle S_z\rangle = -0.82$ (14), i.e., $\mu_L^{Ir}/\mu_S^{Ir} = \langle L_z\rangle/2\langle S_z\rangle = 0.12$ (7), in good agreement with the band structure calculations. Since these experiments have been performed in zero external field, the orientation of μ_S^{Ir} relative to the total Cr magnetic moment (μ^{Cr}) is undetermined. As to the relative direction of the components of the Ir moments, they show that μ_L^{Ir} and μ_S^{Ir} are parallel, confirming the prediction of band structure calculations.

The XMCD measurements were done at the European Synchrotron Radiation Facility located at Grenoble (France) using the spectrometer of beam line 6 [6]. Relative to the only previous measurements at the $L_{II,III}$ edges of Ir [7], our measurements were performed with an undulator source, keeping the direction of the magnetic field fixed while reversing the sign of the circular polarization of the light at the end of each energy scan. In Ref. [7] the field direction was changed every second. The spectra, which were recorded in transmission mode, are presented in Fig. 2a and Fig. 2b. At the two edges we observe stronger white lines that in $Fe_{0.97}Ir_{0.03}$ [7], indicative of a larger $5d$ electron density at the Fermi energy. As found previously for $Fe_{0.97}Ir_{0.03}$, the dichroic signal is of opposite sign at the two edges. Using the first sum rule [8] we deduce $\langle L_z\rangle = 0.12$. This positive value shows that μ_L^{Ir} is antiparallel to μ^{Cr} and confirms the prediction of the band structure calculations. The magnitude of $\langle L_z\rangle$ is also in reasonable agreement with the Mössbauer results and the theory. We remark that the choice of the energy position of the step like function used for the background removal from the measured spectra is at the origin of incertainty in the determination of the magnitude of $\langle L_z\rangle$ but not its sign. Because of our absence of knowledge about the expectation value of the magnetic dipolar operator $\langle T_z\rangle$, we cannot safely determine the spin contribution to the Ir magnetic moment using the second sum rule [9]. The experiment gives $(2\langle S_z\rangle + 7\langle T_z\rangle)/(3n_h) = -0.0387$ where n_h is the number of holes ($n_h = 5$).

Using ^{193}Ir Mössbauer spectroscopy and XMCD measurements at the Ir L_{II} and L_{III} edges we have studied the Ir doped CrO_2. Our results basically confirm predictions based on band structure calculations : μ_S^{Ir} and μ_L^{Ir} are parallel

and μ_L^{Ir} is antiparallel to μ^{Cr}. The magnitude of μ_S^{Ir} and μ_L^{Ir} is in accord with theory. A closed confirmation of the theory would be to the check that the μ_S^{Cr} and μ_S^{Ir} are antiparallel. In principle this can be achieved by performing Mössbauer experiments in a external field.

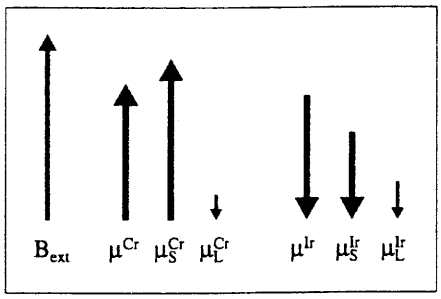

Figure 1: Sketch of the orientation of the different magnetic moments relative to the external field B_{ext}. The Cr moments which account for the bulk of the magnetization are aligned by B_{ext}. Band structure calculations [3] predict the orientation of the different contributions to the Cr and Ir moments as displayed in the figure. The lengths of the arrows do not scale to the magnitude of the different moments. Mössbauer experiments confirm that the coupling between μ_S^{Ir} and μ_L^{Ir} is parallel. XMCD experiments confirm the orientation of μ_L^{Ir} relative to μ^{Cr}.

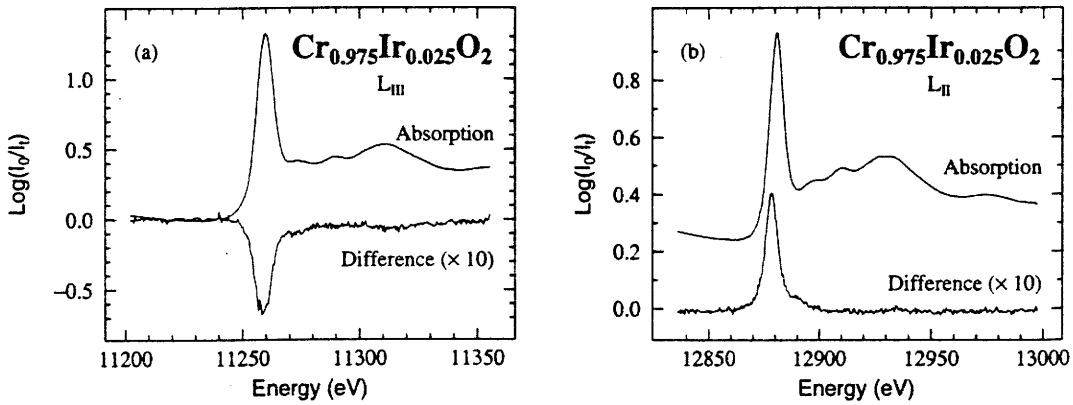

Figure 2a (left hand side): Absorption spectrum γ_+ and dichroic asymmetry spectrum $\Delta\gamma$ ($\Delta\gamma = \gamma_+ - \gamma_-$) measured at the L_{III} edges of iridium in $Cr_{0.975}Ir_{0.025}O_2$. The intensity of the applied magnetic field was 1 T and the temperature 100 K. The index + (−) specifies that the applied field is parallel (antiparallel) to the X-ray helicity.
Figure 2b (right hand side): same caption as for Fig. 2a but for the L_{II} edge.

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